Analysis of self-trapping using the wave equation with high-order nonlinear electric permittivity*

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The self-trapping of light pulses is considered using the classical nonlinear wave equation of Chiao, Garmire, and Townes, modified to include nonlinearities up to sixth order in electric field. A phenomenological approach is adopted in which the nonlinear coefficients are considered without reference to a specific molecular self-trapping mechanism. It is found that filament formation can occur only over a narrow range of values of the nonlinear coefficients. The method can be used as a criterion to test the validity of various self-trapping mechanisms.

Self-focusing and self-trapping of giant light pulses traversing a transparent medium are governed by the wave equation with a field-dependent refractive index. The exact form of the nonlinearities depends on the details of the specific molecular processes which give rise to filament formation. Many processes, some very sophisticated and unusual, have been invoked in order to account for the observed behavior.

In order to identify the molecular processes which play an important role in self-trapping, it is important to study the behavior of a light beam in a nonlinear medium without appeal to any particular mechanism of nonlinearity. Such a phenomenological treatment, based on the classical nonlinear wave equation, was initiated by Chiao, Garmire, and Townes in 1964.[1] A different approach has been given by Kelley and by Javan and Kelley,[2] by Akhmanov *et al.*,[3] by Maier *et al.*[4] and by others, where self-focusing rather than self-trapping is discussed.

In the present paper we extend the method of Chiao *et al.* to the case where nonlinearities higher than order two are included.[5] As early as 1966[6] it was recognized that it is necessary to consider nonlinearities higher than $\epsilon_2 E^2$ in order to obtain some knowledge of the diameter of a self-trapped beam or light filament. In Ref. 5 a nonlinear electric permittivity of the following form was proposed to describe the formation of filaments in liquids and isotropic solids:

$$\epsilon = \epsilon_0 + \epsilon_2 E^2 + \epsilon_4 E^4 + \epsilon_6 E^6 , \tag{1}$$

where $\epsilon_0 = n_0^2$ is the electric permittivity of the medium at the optical frequency in the absence of an electric field and ϵ_2, ϵ_4, and ϵ_6 are nonlinear coefficients. E is the external electric-field intensity expressed in this paper in electrostatic units. In the case where E is the electric field of a powerful laser beam traversing a liquid or solid medium, and if the relaxation time associated with the response to this field is slow compared to the inverse optical frequency, then the time averaged form of Eq. (1) should be used:

$$\epsilon_{\text{eff}} = \epsilon_0 + \tfrac{1}{2}\epsilon_2 E_0^2 + \tfrac{3}{8}\epsilon_4 E_0^4 + \tfrac{5}{16}\epsilon_6 E_0^6 , \tag{2}$$

where E_0 is the amplitude of E and $\epsilon_{\text{eff}} = n_{\text{eff}}^2$.

In what follows we utilize the wave equation of Chiao, Garmire, and Townes to which two higher-order nonlinear terms with coefficients α and β are added:

$$\frac{d^2E^*}{dr^{*2}} + \frac{1}{r^*}\frac{dE^*}{dr^*} - E^* + E^{*3} + \alpha E^{*5} + \beta E^{*7} = 0 . \tag{3}$$

Here $E^* = E^*(r^*)$ is the dimensionless amplitude of $E(r) = E_0(r)\cos(k_z z - \omega t)$, and r and r^* are the actual and dimensionless radial distances from the filament axis, respectively. The nonlinear coefficients α and β are given by

$$\alpha = \frac{3}{4}\frac{\epsilon_4}{\epsilon_2}\left(\frac{E_0}{E^*}\right)^2 , \tag{4}$$

$$\beta = \frac{5}{8}\frac{\epsilon_6}{\epsilon_2}\left(\frac{E_0}{E^*}\right)^4 = \frac{10}{9}\frac{\epsilon_2\epsilon_6}{\epsilon_4^2}\alpha^2 . \tag{5}$$

The basic procedure for calculating the eigenfunctions $E^*(r^*)$ for cylindrical filaments is the same as in Refs. 1 and 5, and the paper of Gustafson *et al.*[7] The dimensionless distance r^* is defined as

$$r^* = \Gamma r , \tag{6}$$

with

$$\Gamma = (k_z^2 - k^2)^{1/2} , \tag{7}$$

where k_z, the propagation constant along the beam

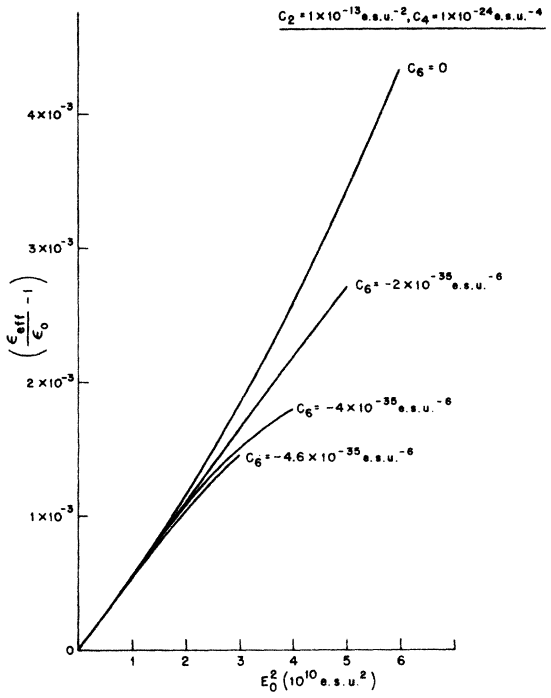

FIG. 1. Relative increase of $\epsilon_{\text{eff}} = n_{\text{eff}}^2$ as a function of E_0^2 for $C_2 = 1\times10^{-13}$ esu^{-2}, $C_4 = 1\times10^{-24}$ esu^{-4}, and different values of C_6.

axis (z axis), is given by

$$k_z = n_{\text{eff}}\,\omega/c \tag{8a}$$

and

$$k = n_0\omega/c\,. \tag{8b}$$

The dimensionless electric field amplitude is defined as

$$E^*(r^*) = b^{1/2}E_0(r)\,, \tag{9}$$

where

$$b = \tfrac{1}{2}\epsilon_2(k_0^2/\Gamma^2)\,, \tag{10}$$

with

$$k_0 = \omega/c\,, \tag{11}$$

so that

$$\Gamma = k_0\epsilon_0^{1/2}(\epsilon_{\text{eff}}/\epsilon_0 - 1)^{1/2}\,. \tag{12}$$

The aim of the calculation is to obtain the diameter of the self-trapped filament as a function of the power P fed into the beam. For a given set of parameters ϵ_0, ϵ_2, ϵ_4, and ϵ_6, which characterize the nonlinearity of a medium, we plot the quantity

$$\epsilon_{\text{eff}}/\epsilon_0 - 1 = \tfrac{1}{2}C_2E_0^2 + \tfrac{3}{8}C_4E_0^4 + \tfrac{5}{16}C_6E_0^6\,, \tag{13}$$

with

$$C_2 = \epsilon_2/\epsilon_0,\quad C_4 = \epsilon_4/\epsilon_0\,,\quad C_6 = \epsilon_6/\epsilon_0\,, \tag{14}$$

as a function of E_0^2. Two such series of curves are plotted in Figs. 1 and 2 for various values of the parameters C_2, C_4, and C_6. The segments of the curves shown correspond to the approximate intervals where convergent solutions of Eq. (3) exist. Examples of such solutions are displayed in

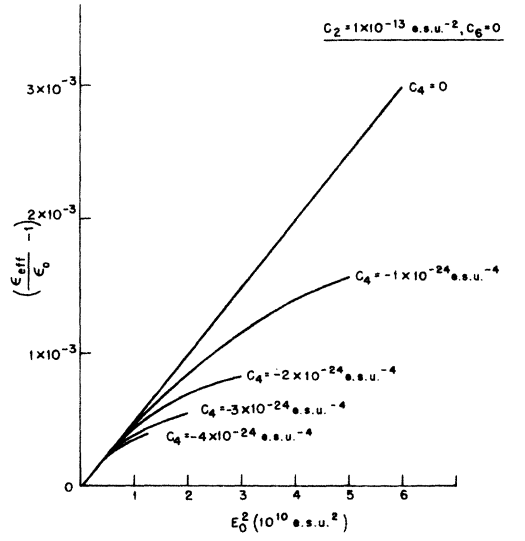

FIG. 2. Relative increase of $\epsilon_{\text{eff}} = n_{\text{eff}}^2$ as a function of E_0^2 for $C_2 = 1\times10^{-13}$ esu^{-2}, $C_6 = 0$, and various values of C_4.

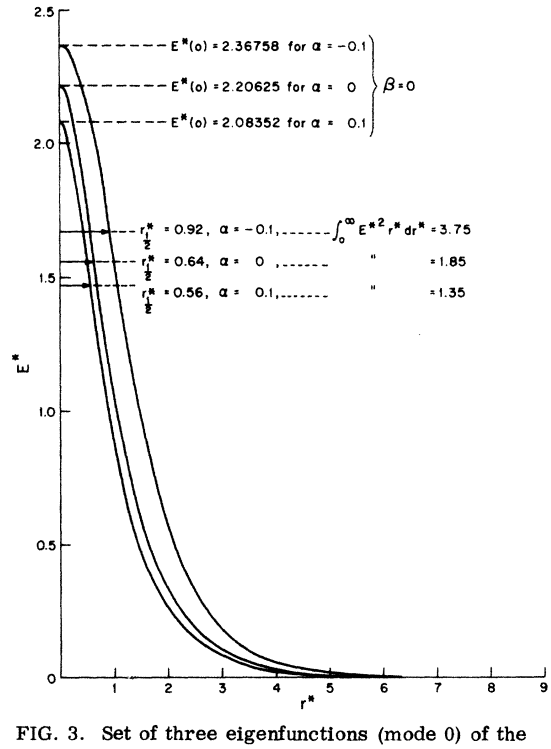

FIG. 3. Set of three eigenfunctions (mode 0) of the nonlinear wave equation (3) for $\alpha = -0.1$, 0, $+0.1$, with $\beta = 0$. The half-intensity radii $r^*_{1/2}$ are shown.

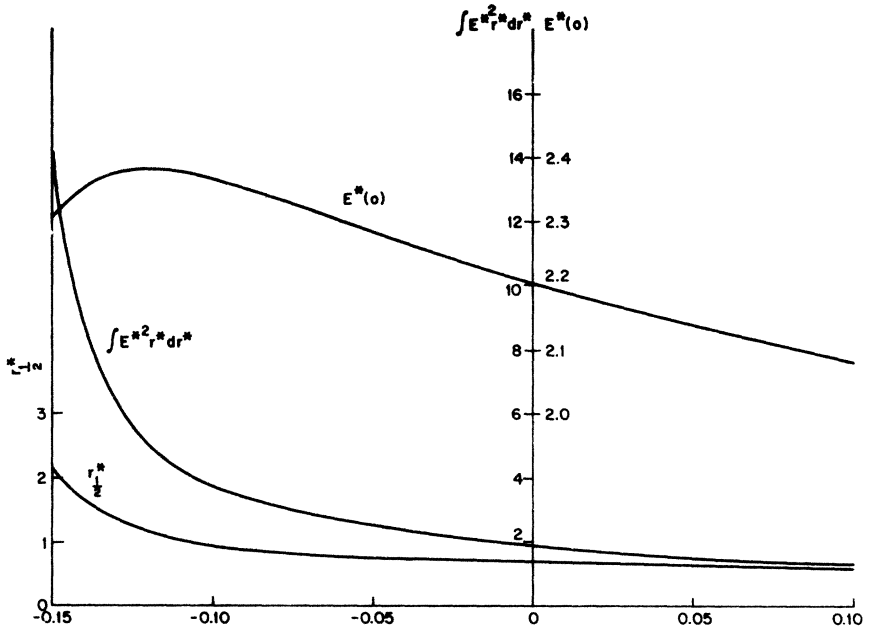

FIG. 4. Electric-field intensity $E^*(0)$ on the axis of the filament, half-intensity radii $r^*_{1/2}$ and $\int_0^\infty E^{*2} r^* dr^*$ as functions of α, with $\beta = 0$. All quantities are expressed in dimensionless units.

Fig. 3, where three eigenfunctions are plotted for the cases $\alpha = -0.1$, 0, $+0.1$, and $\beta = 0$. For each curve we show $E^*(0)$, the value of the field amplitude on the axis of the filament, and $E^*(r^*_{1/2})$, its value at the point $r = r^*_{1/2}$, where the light intensity E^{*2} drops to half its maximum value.

The power P fed into the beam is given by

$$P = \frac{c\lambda^2}{16\pi^2 n_2} \int_0^\infty E^{*2} r^* dr^* . \tag{15}$$

This integral has been evaluated for different values of α and β. In Fig. 4 the quantities $\int_0^\infty E^{*2} r^* dr^*$, $E^*(0)$, and $r^*_{1/2}$ are plotted as functions of α for $\beta = 0$.

In order to calculate the filament diameter D, defined as twice the diameter at the half-maximum intensity point, particular values for the constants C_2, C_4, and C_6 must be chosen. For different values of α and β the eigenfunctions of Eq. (3) are computed numerically. $E^*(0)$, $\int_0^\infty [E^*(r^*)]^2 r^* dr^*$, and $r^*_{1/2}$ can then be calculated from these eigenfunctions. From the computed value of E^*, E_0 can

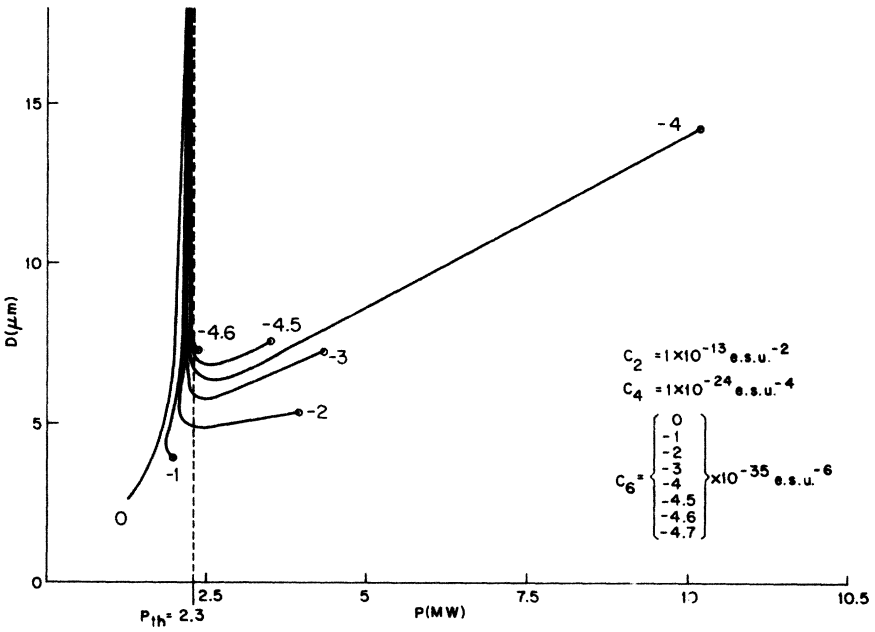

FIG. 5. Filament diameter D in μm as a function of the power P in MW fed into the beam for the set of C coefficients shown in Fig. 1.

be obtained using Eq. (4). Γ can then be obtained from Eqs. (2) and (12) and from this $r_{1/2}$ can be obtained by means of Eq. (6). The filament diameter D is defined as $D = 2D_{1/2} = 4r_{1/2}$. The power fed into the beam can be calculated from $\int_0^\infty E^{*2} r^* dr^*$. In this way plots of D as a function of P are obtained.

Two families of curves of the functions $D(P)$ calculated for $n_0 = 1.5$ and $\lambda_0 = 0.7$ μm, are plotted in Figs. 5 and 6 using the dependence of $(\epsilon_{\text{eff}}/\epsilon_0) - 1$ on E_0^2 shown in Figs. 1 and 2, respectively. The reasonable value of 1×10^{-13} esu is chosen for C_2, leading to minimum filament diameters of between 4 and 15 μm, in good agreement with experiments for various liquids. Figure 5 ($C_4 > 0, C_6 < 0$) corresponds to the case where the nonlinearity is due to the optical Kerr effect, where anisotropic molecules are oriented in the electric field of a light wave.[8] In this case self-trapping starts from slightly below threshold (P_{th}, Fig. 5), whereas in the case shown in Fig. 6 ($C_4 < 0$), it starts from immediately above threshold.

In the case shown in Fig. 5 the eigenfunctions exhibit very peculiar behavior. Above certain values of the power P there are no converging solutions of Eq. (3), and consequently not all the power can be trapped in a single filament. This situation corresponds to the end points of the curves in Fig. 5. Beyond these points only part of the available power can be fed into a single filament, and it is plausible that under these conditions many filaments could be formed. In contrast, the curves of Fig. 6 are all continuous in the power region of interest, and no such end points occur until power levels of many hundreds of MW are reached.

Another peculiarity of the case considered in Fig. 5 is that as increasing power is fed into the beam the filament formation starts immediately with a very small diameter of a few μm. This is seen in Fig. 5 for different values of C_6, and also in Fig. 2 of Ref. 5 for different values of C_4. At the minimum value of the power at which the filament starts, which is somewhat smaller than the threshold power P_{th}, there exist two types of solutions. In the first type the filament diameter rapidly diverges with increasing power. This type of solution is nonstationary. In the second type the filament diameter tends to a minimum value as is actually observed in the experiments.

The importance of the C_2 coefficient in the process of filament formation can be seen in Fig. 7 for the case $C_4 > 0$, $C_6 < 0$, corresponding to a nonlinearity due to orientation, and in Fig. 8 for the case of $C_4 < 0$. Evidently, in both cases C_2 must be positive, and with increasing values of C_2 both the filament diameter and the threshold power decrease.

It is important to note that converging solutions exist only over a rather narrow range of values of the nonlinear coefficients. Thus, the order of magnitude of these coefficients expressed in esu, should be

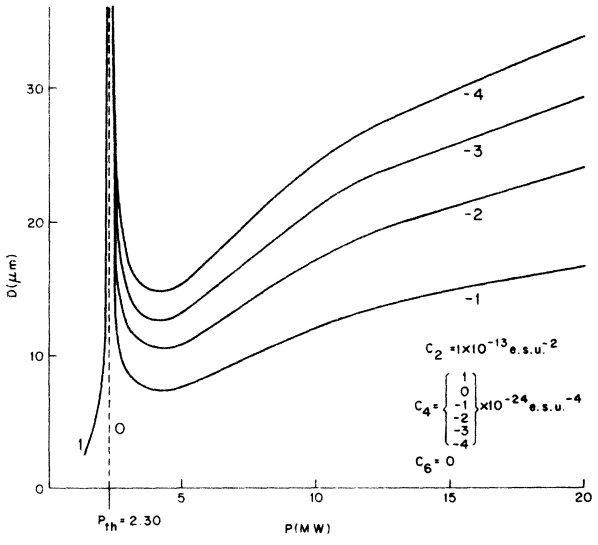

FIG. 6. Filament diameter D in μm as a function of the power P in MW for the set of C coefficients shown in Fig. 2.

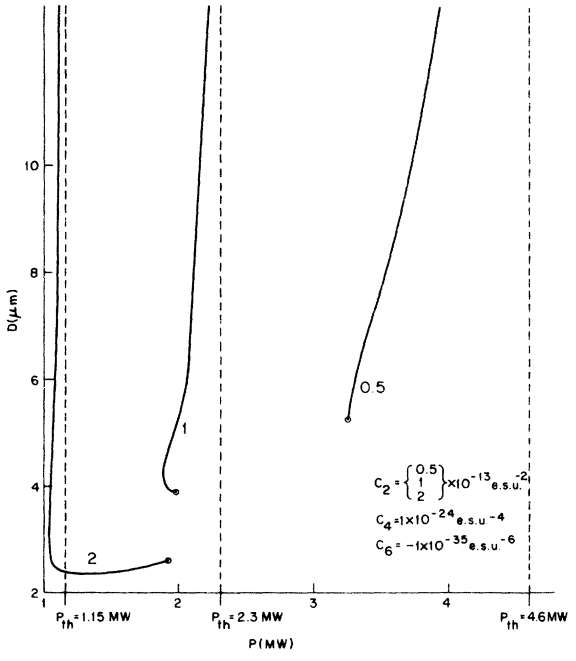

FIG. 7. Filament diameter D in μm as function of the power P in MW for different values of C_2, with $C_4 = 1 \times 10^{-24}$ esu^{-4} and $C_6 = -1 \times 10^{-35}$ esu.

$C_2 \cong 10^{-13}$,

$C_4 \cong 10^{-24}$,

$C_6 \cong -10^{-35}$

for the case $C_4 > 0$, and

$C_2 \cong 10^{-13}$,

$C_4 \cong -10^{-24}$

for the case $C_4 < 0$.

We may also look at the self-trapping effect in a quite different way, viz., by assuming a particular molecular mechanism for self-trapping, which gives rise to a nonlinear refractive index of a particular form. By expanding the corresponding ϵ in power of E^2, one then obtains a set of coefficients which may be compared with those found in our phenomenological treatment. This could constitute a criterion to test the validity of any particular mechanism of interest.

Free orientation of anisotropic molecules as a self-trapping mechanism has been ruled out in this way. This mechanism is only possible if less than 2% of the molecules exhibit this type of behavior.[8] Nevertheless, the very ingenious recent experiments of Reintjes and Carman[9] in fact do point to molecular orientation as the dominant mechanism of self-trapping in different Kerr liquids.

How can the effect of molecular orientation undergo such a drastic reduction? The answer to this question was given by Gustafson and Townes,[10]

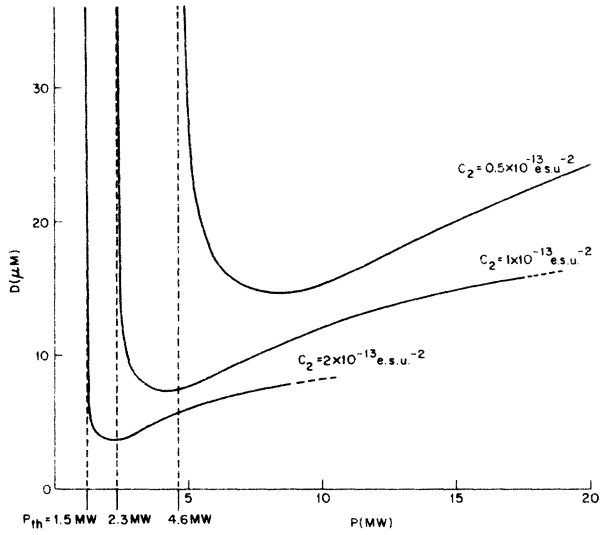

FIG. 8. Filament diameter D as a function of the power P in MW for different values of C_2, with $C_4 = -1\times10^{-24}$ esu^{-4} and $C_6 = 0$.

who showed that the nonlinear response to internal pressure reduces the orientation effect by a factor of 10 in liquid CS_2. Quite recently it was pointed out[11] that another factor-of-10 reduction was expected because of the interaction between dipoles induced by the electric field of the light wave,[12] resulting in enhancement of the lattice vibrations.

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